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CONVENTION

607174

APPLICATION FOR A STANDARD PATENT

☒ We Charles of the Ritz Group Ltd.

of 40 West 57th Street,
New York,
New York,
UNITED STATES OF AMERICA.

APPLICATION ACCEPTED AND AMENDMENTS

ALLOWED

4.12.90

hereby apply for the grant of a standard patent for an invention
entitled:

ULTRAVIOLET ABSORBING COMPOUNDS AND FORMULATIONS INCLUDING
SAME

which is described in the accompanying complete specification.

Details of basic application

Number of basic application: 947,728

Convention country in which
basic application was filed: UNITED STATES OF AMERICA

Date of basic application : 30 December 1986

Address for Service:

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29 DEC 1987

Melbourne

Dated: 28 December 1987

PHILLIPS ORMONDE & FITZPATRICK
Attorneys for:
Charles of the Ritz Group Ltd.

By

Our Ref : 79850

POF Code: 1431/47599

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185
MAIL OFFICER: P ATTACHED

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DECLARATION FOR A PATENT APPLICATION

INSTRUCTIONS

(a) Insert "Convention" if applicable
(b) Insert FULL name(s) of applicant(s)

(c) Insert "of addition" if applicable
(d) Insert TITLE of invention

(e) Insert FULL name(s) AND address(es) of declarant(s) (See heading*)

(f) Insert FULL name(s) AND address(es) of actual inventor(s)

(g) Rights how applicant(s) derive(s) title from actual inventor(s) (See heading*)

(h) Insert country, filing date, and basic application(s) for the EACH basic application

(i) Insert PLACE of signing

(j) Insert DATE of signing

(k) Signature(s) of declarant(s)

Note: No legalization or other witness required

In support of the (a) convention application made by

(b) CHARLES OF THE RITZ GROUP, LTD. A corporation of the State of Delaware, U.S.A., 40 West 57th Street, New York, New York, 10019 (U.S.A.)

(hereinafter called "applicant(s)" for a patent (c) for an invention entitled (d) ULTRAVIOLET ABSORBING COMPOUNDS AND FORMULATIONS INCLUDING SAME

I/We (e) Patrick Boland, Executive Secretary of Charles of the Ritz Group, Ltd. of 40 West 57th Street, New York, New York, 10019 United States of America

do solemnly and sincerely declare as follows:

1. ~~XXXXXX~~ (or, in the case of an application by a body corporate)
1. I am/We are authorized to make this declaration on behalf of the applicant(s).
2. ~~XXXXXX~~
2. (f) Arthur C.W. Georgalas
12 Bellevue Avenue
Leonardo, New Jersey 07737, U.S.A.

is/are the actual inventor(s) of the invention and the facts upon which the applicant(s) is/are entitled to make the application are as follows:

- (g) Applicant is the assignee of the invention from the actual inventor.

(Note: Paragraphs 3 and 4 apply only to Convention applications)

3. The basic application(s) for patent or similar protection on which the application is based is/are identified by country, filing date, and basic application(s) as follows:

(h) U.S.A. U.S. Serial No. 947,728, filed Dec. 30, 1986
Arthur C.W. Georgalas

4. The basic application(s) referred to in paragraph 3 hereof was/were the first application(s) made in a Convention country in respect of the invention the subject of the application.

Declared at (i) New York, New York, U.S.A.

Dated (j) January 21, 1988

(k) CHARLES OF THE RITZ GROUP, LTD.

By: 
Patrick Boland
Assistant Secretary

To: The Commissioner of Patents

P15/78

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ULTRAVIOLET ABSORBING COMPOUNDS AND FORMULATIONS INCLUDING SAME

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(56) Prior Art Documents
US 4683134
GB 1033299
FR 2520615

(57) Claim

1. A substantive ultraviolet absorbing sunscreen or sunblock composition consisting essentially of one or more ultraviolet absorbing compounds having an acid group, a fatty compound selected from the group consisting of fatty amines, primary ether fatty amines, fatty amidopropyl dimethylamines, fatty amidoethyl diethylamines, fatty amidoethyl ethanolamines, cocyl imidazoline and alkyl dimethylamines and optionally one or more emollients, emulsifiers, preservatives, antioxidants, fragrances, coloring agents or chelating agents, wherein said fatty compound is present in an amount effective to provide enhanced substantivity to said ultraviolet absorbing compounds.

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COMPLETE SPECIFICATION
(ORIGINAL)

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amendments made under
Section 49 and is correct for
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Related Art:

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Name(s) of Applicant(s):

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Complete Specification for the invention entitled:

ULTRAVIOLET ABSORBING COMPOUNDS AND FORMULATIONS INCLUDING SAME

Our Ref : 79850
POF Code: 1421/47599

The following statement is a full description of this invention, including
the best method of performing it known to applicant(s):

5 The present invention relates to improved ultraviolet absorbing compounds and more particularly concerns sunscreen or sunblock formulations which include such compounds and which have enhanced substantivity.

 Sunscreen and sunblock formulations provide excellent protection against severe sun burning of exposed skin. These formulations include agents which can absorb ultraviolet light at various wavelengths.
10 For example, ultraviolet radiation from the sun reaching the earth's atmosphere is typically divided into two ranges; the UV-A range (wavelengths of 320 to 400 nanometers) and the UV-B (wavelength of 290 to 320 nanometers). Numerous UV-A and UV-B absorbing
15 compounds or agents are known in the art.

 Since different degrees of energy are absorbed by skin exposed to these varying wavelengths of ultraviolet radiation, sunscreen preparations can be formulated to extend the period of time it takes the
20 sun to produce a sunburn by varying the amounts of the UV-A and UV-B absorbers incorporated therein.

 However, many commercially available sunscreen or sunblock formulations suffer from the disadvantage that they are washed off the skin during
25 bathing in pool water or ocean water. This results in certain areas of the skin being exposed to greater amounts of ultraviolet radiation than intended.

 Improved ultraviolet absorbing compounds resistant to water wash-off, i.e. having enhanced
30 substantivity, would be a useful addition to the art.

~~In accordance with the present invention an improved ultraviolet absorbing compound, and a sunscreen or sunblock formulation including such a compound, are provided. The improved ultraviolet~~



In accordance with the present invention there is provided a substantive ultraviolet absorbing sunscreen or sunblock composition consisting essentially of one or more ultraviolet absorbing compounds having an acid group, a fatty compound selected from the group consisting of fatty amines, primary ether fatty amines, fatty amidopropyl dimethylamines, fatty amidoethyl diethylamines, fatty amidoethyl ethanolamines, cocyl imidazoline and alkyl dimethylamines and optionally one or more emollients, emulsifiers, preservatives, antioxidants, fragrances, coloring agents or chelating agents, wherein said fatty compound is present in an amount effective to provide enhanced substantivity to said ultraviolet absorbing compounds.



~~absorbing compound comprises, in combination, an~~
~~ultraviolet absorbing agent having an acid group and a~~
~~fatty amine.~~ The resulting improved ultraviolet
absorbing compound and formulations into which it is
incorporated have enhanced substantivity and thereby
greater water resistance.

Further in accordance with the present
invention there is provided a sunscreen or sunblock
formulation comprising one or more ^{ultraviolet} ~~ultraviolet~~ absorbing
agents, which agents include an acid group, in
combination with a fatty amine, water and optionally
one or more emollients, emulsifiers, preservatives,
antioxidants, fragrances, coloring agents or chelating
agents.

It has been found that sunscreen or sunblock
formulations are provided with greater substantivity
when they are formulated to include ultraviolet
absorbing compounds having an acid group and one or
more fatty amines. In fact, the reaction products of
the acid-containing UV absorbers and fatty amines also
possess this enhanced substantivity. Therefore, these
reaction products represent improved ultraviolet
absorbing compounds and, as such, are considered part
of the present invention.

In one embodiment, a sunscreen or sunblock
formulation according to the present invention includes
compounds which have an acid group and which absorb
light in the UV-A (320-400 nm) and UV-B (290 to 320 nm)
regions, one or more fatty amines and typically also
contains water, emollients, emulsifiers, thickeners,
preservatives, coloring agents, fragrances,
antioxidants and the like.

The formulation of the present invention is
preferably an oil-in-water type emulsion since this



type of emulsion affords a better cosmetic feel to the product. However, the product may also be formulated as a water-in-oil emulsion, cream base or oil base. Depending upon the choice of ingredients, the composition has a semi-solid, cream-like consistency which can be packaged in a plastic squeeze tube or it has a lotion type consistency which can be packaged in a plastic squeeze container.

As mentioned above, the present invention relies on the combination of a UV absorbing agent having an acid group, with a fatty amine. The fatty amine may be a primary, secondary or tertiary amine derived from, for example, tallow, coconut oil, soybean oil, marine oils, rapeseed and the like. It can also be a primary ether amine, i.e. a fatty amine derived from an alcohol instead of from a fatty acid. The fatty amine can also be a lipophilic derivative of a fatty amidopropyl dimethylamine base.

Suitable fatty amines include lauramidopropyl dimethylamine (Lexamine L-13), oleamidopropyl dimethylamine (Lexamine O-13), ricinoleamidopropyl dimethylamine (Lexamine R-13), stearamidoethyl ethanolamine (Chemical base 39), stearamidoethyl diethylamine (Chemical base 6532), cocoyl imidazoline (Miramine CC), and alkyldimethylamine (ADMA²).

In addition to the fatty amines, other amines or bases may be employed to enhance the solubility of the UV-absorbers in the present formulations. These include, but are not limited to, water-soluble amines (e.g. triethanolamine), ammonia or alkali hydroxides.

In accordance with the present invention the fatty amine is typically combined with both the UV-A and UV-B absorbers having acid groups. Alternatively, the fatty amine can be combined with one or the other

of the UV absorbers. The improved UV absorbing compound of the present invention may comprise the fatty amine in a molar ratio to the UV absorbers (A and/or B) between about 0.25:1 and 2:1 and preferably from about 0.5:1 to 1.2:1.

5 A sunscreen or sunblock formulation according to the present invention may comprise between about 0.5 and 20 percent by weight, and preferably includes between about 1 and 8 percent by weight of the fatty amines.

10 Suitable UV-A absorbers having acid groups include sulisobenzone (2-hydroxy-4-methoxybenzophenone-5-sulfonic acid; Uvinul MS-40), thiocetic acid (lipoic acid), and the like. The sulisobenzone is the preferred UV-A absorber. The desired UV-A absorber
15 will be present in the final product in an amount of from about 0.25 to about 20 percent by weight, and preferably in an amount of from about 1 to about 8 percent by weight of the total formulation. The amount
20 will vary according to the particular absorber selected and whether the formulation is intended to prevent, minimize or permit tanning.

25 Suitable UV-B absorbers having acid groups include 2-phenylbenzimidazole-5-sulfonic acid (Eusolex[®] 232), p-aminobenzoic acid, salicylic acid, methoxycinnamic acid, cinnamic acid, ferrulic acid and the like. The Eusolex[®] 232 is the preferred UV-B
30 absorber. The desired UV-B absorber will be present in the final product in an amount of from about 0.25 to about 20 percent by weight, and preferably from about 1 to 8 percent by weight of the total formulation. The amount will vary according to the particular absorber selected and whether the formulation is intended to prevent, minimize or permit tanning.

The total amount of UV absorbers included within the formulation will be between about 0.5 and 20 percent by weight; which amount will determine whether the formulation is a sunblock or a sunscreen.

5 In addition to the fatty amine and UV absorbers, the present formulation can include from about 60 to about 95 percent by weight of a solvent, e.g. ethanol, isopropanol, etc.

10 Alternatively, in addition to the fatty amine and UV absorbers, the formulation may contain from about 50 percent to about 90 percent and preferably from about 60 to about 80 percent by weight of water, from about 1 percent to about 20 percent and preferably from about 1 to about 5 percent by weight of emollients, from about 1 percent to about 10 percent and preferably from about 1 to about 5 percent by weight of emulsifiers, from about 0.05 to about 2 percent and preferably from about 0.1 to about 1 percent by weight of preservatives and antioxidants, and less than about 1 percent by weight of fragrance, coloring agents and chelating agents (such as EDTA).

20 Suitable emollients include mineral oil, avocado oil, squalane, octyl palmitate, cocoa butter, sesame oil, petrolatum, propylene glycol dicaprylate/dicaprate, isopropyl myristate, dimethicone (e.g. Silicon 225), etc. The formulation will preferably contain a mixture of several of these emollients or others which are approved for cosmetic use.

30 Suitable emulsifiers include polyethylene glycol 20 sorbitan monolaurate (Polysorbate 20), glyceryl stearate, polyethylene glycol 100 stearate, polyethylene glycol 20 stearyl ether (Brij 78, Steareth 20), polysorbate 80 (Tween 80), etc. The

formulation will preferably contain a mixture of two or more of these emulsifiers or others which are approved for cosmetic use.

Suitable preservatives include imidazolidinyl urea (Germall 115), methylparaben (Tegosept M), quaternium-15 (N-(3-chloroallyl)hexaminium chloride, Dowicil 200), propylparaben (Tegosept P), dimethyldimethoyl hydantoin (Glydant), benzyl alcohol and/or phenoxyethanol, etc., and a preferred antioxidant is a mixture of butylated hydroxyanisole, propylene glycol, propyl gallate and citric acid (Tenox 2). The formulation will preferably contain the antioxidant mixture and one or more of the preservatives or any other preservatives and antioxidants approved for cosmetic use. *

As discussed above, by varying the percentage of ingredients the formulation can be obtained in a lotion or semi-solid form. For example, in formulating the product as a lotion, water would be included at from about 60 percent to 65 percent by weight of the final product and one or more humectants such as propylene glycol, glycerin, 1,3-butylene glycol, sorbitol, polyethylene glycols (for example, Carbowax 400), could be included at up to about 20 percent by weight of the final product.

The composition of the invention will optionally include a thickener in an amount within the range of from about 0.05 to about 1 percent and preferably from about 0.05 to about 0.3 percent by weight. Examples of thickeners which may be employed herein include, but are not limited to, stearic acid, magnesium aluminum silicate, stearoxydimethicone, hydroxyethyl cellulose, hydroxypropyl cellulose or xanthan gum.

Skin conditioning agents which may optionally be present in the composition of the invention include allatoin, d- or dl-panthenol, hydrolyzed animal protein and the like. Such conditioning agents may be present in an amount within the range of from about 0.01 to about 5 percent and preferably from about 0.05 to about 2 percent by weight depending upon the ultimate use of the skin preparation.

The process techniques will vary depending upon the particular ingredients employed. For example, in one such process a first aqueous blend can be prepared by mixing the UV absorbing agents, a water soluble organic base such as triethanolamine, a chelating agent such as EDTA and other water soluble ingredients, if desired, into the deionized water. This is mixed thoroughly and typically heated, for example, to about 70°C. The fatty amine is added, by itself or with an alcohol, to complete the aqueous blend. A second blend is prepared by mixing an emollient, e.g. dimethicone emulsion, with a small amount of deionized water and one or more emulsifiers, if desired. The first and second blends were sweep mixed at room temperature to form an emulsion. A third blend comprising preservatives and other desired ingredients in a small amount of deionized water is thereafter added to the emulsion at room temperature to complete the desired formulation.

A preferred sunblock formulation in accordance with the present invention will include about 70 percent by weight of water, about 4 percent by weight of Eusolex® 232, about 4.5 percent by weight of triethanolamine, about 5 percent by weight of methoxycinnamic acid, about 2.5 percent by weight of salicylic acid, about 1 percent by weight of

5 sulisobenzone, about 4 percent by weight of cocoyl
imidazoline, about 3 percent by weight of emollients,
about 5 percent by weight of emulsifiers, about 0.35
percent by weight of thickener, and up to 1 percent by
weight of combined preservatives, antioxidants,
6 fragrances and chelating agents.

10 A preferred sunscreen formulation which
protects, but still permits gradual tanning, according
to the present invention includes about 80 percent by
weight of water, about 4 percent by weight of Eusolex®
232, about 1 percent by weight of sulisobenzone,
about 6 percent by weight of cocoyl imidazoline,
about 3 percent by weight of emollients, about 5 percent
by weight of emulsifiers, about 0.35 percent by weight
of thickener and up to about 1 percent by weight of
15 combined preservatives, antioxidants, fragrances, and
chelating agents.

20 The present invention will be further described by
reference to the following Examples. It should be
understood, however, that the present invention is not
to be limited to the details described therein.

25

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Example 1

A sunblock formulation in the form of a lotion having the following composition was prepared as described below.

5	Ingredient	Percent by Weight
	<u>Blend I</u>	
	Deionized Water	70.0
	Eusolex® 232 (UV-B absorber)	4.0
10	Methoxycinnamic acid (UV-B absorber)	5.0
	Salicylic acid (UV-B absorber)	2.5
	Sulisobenzene (Uvinul MS-40; UV-A absorber)	1.0
15	Triethanolamine (for solubility of UV absorbers)	4.5
	Cocoyl imidazoline (Miramine CC; fatty amine)	4.0
20	EDTA (chelating agent)	0.10
	<u>Blend II</u>	
	Dimethicone emulsion (Silicon 225; emollient)	5.0
25	Deionized water	5.0
	<u>Blend III</u>	
	Germall II (preservative)	0.2
	Glydant (preservative)	0.3
30	Deonized water	1.0

Aqueous blend I was prepared by mixing the UV absorbers, triethanolamine, EDTA and fatty amine into

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Blend I

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Blend II

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Blend III

Germall II (preservative)	0.2
Glydant (preservative)	0.3
Deionized water	1.0

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Example 3

A sunscreen formulation in the form of a lotion having the following composition was prepared as described below.

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Ingredient	Percent by Weight
------------	-------------------

Blend I

15	Eusolex® 232 (UV-B absorber)	4.0
	Sulisobenzone (Uvinul MS-40; UV-A absorber)	2.0
	Stearamidoethyl diethylamine (Chemical base 6532; fatty amine)	8.0
20	Ethanol	86.0

20

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The above ingredients were heated to about 70°C and mixed thoroughly. Upon cooling to room temperature the mixture formed the lotion of the present invention. Upon evaporation of the alcohol a viscous yellow paste was formed which was found to be a water-dispersible substantive UV-absorbing compound according to the present invention, which could, in turn, be used in other formulations.

30

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A substantive ultraviolet absorbing sunscreen or sunblock composition consisting essentially of one or more ultraviolet absorbing compounds having an acid group, a fatty compound selected from the group consisting of fatty amines, primary ether fatty amines, fatty amidopropyl dimethylamines, fatty amidoethyl diethylamines, fatty amidoethyl ethanolamines, cocyl imidazoline and alkyl dimethylamines and optionally one or more emollients, emulsifiers, preservatives, antioxidants, fragrances, coloring agents or chelating agents, wherein said fatty compound is present in an amount effective to provide enhanced substantivity to said ultraviolet absorbing compounds.
2. The compound of claim 1 wherein said acid-containing ultraviolet absorbing agents are selected from the groups consisting of UV-A absorbing agents including sulisobenzene and thiocctic acid, and UV-B absorbing agents including 2-phenylbenzimidazole-5-sulfonic acid, salicyclic acid, methoxycinnamic acid, cinnamic acid and furrulic acid.
3. The compound of claim 1 or 2 wherein said fatty amine is selected from the group consisting of lauramidopropyl dimethylamine, oleamidopropyl dimethylamine, ricinoleamidopropyl dimethylamine, steramidoethyl ethanolamine, stearamidoethyl diethylamine, cocyl imidazoline, and alkyl dimethylamine.
4. The compound of claim 1, 2 or 3 wherein the molar ratio of the fatty amine to the absorbing agent including the acid group is from 0.25:1 to 2:1.
5. The compound of claim 4 wherein the molar ratio of the fatty amine to the absorbing agent including the acid group is from 0.5:1 to 1.2:1.
6. A sunscreen or sunblock formulation comprising the substantive ultraviolet absorbing compounds of claim 1 in combination with a solvent.
7. The formulation of claim 6 wherein said solvent is an alcohol.

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8. A substantive ultraviolet absorbing composition consisting essentially of one or more ultraviolet absorbing compounds having an acid group, water and a fatty compound selected from the group consisting of fatty amines, primary ether fatty amines, fatty amidopropyl dimethylamines, fatty amidoethyl diethylamines, fatty amidoethyl ethanolamines, cocyl imidazoline and alkyl dimethylamines and optionally one or more emollients, emulsifiers, preservatives, antioxidants, fragrances, coloring agents or chelating agents, wherein said fatty compound is present in an amount effective to provide enhanced substantivity to said ultraviolet absorbing compounds.



~~and a fatty amine or a compound derived therefrom, wherein said fatty amine or derivative therefrom is not in combination with a fatty acid and is present in an amount effective to provide enhanced substantivity to said ultraviolet absorbing agents, and optionally one or more emollients, emulsifiers, preservatives, antioxidants, fragrances, coloring agents or chelating agents.~~

9. The formulation of claim 8 wherein said one or more ultraviolet absorbing agent are selected from the group of UV-A absorbing agents consisting of sulisobenzone and thiocetic acid and one or more ultraviolet absorbing agents selected from the group of UV-B absorbing agents consisting of 2-phenylbenzimidazole-5-sulfonic acid, salicylic acid, methoxycinnamic acid, cinnamic acid and ferrulic acid.

10. The formulation of claim 8 or 9 wherein said fatty amines are selected from the group consisting of lauramidopropyl dimethylamine, oleamidopropyl dimethylamine, ricinoleamidopropyl dimethylamine, stearamidoethyl ethanolamine, stearamidoethyl diethylamine, cocoyl imidazoline, and alkyldimethylamine.

11. The formulation of claim 8, 9 or 10 comprising from 0.25 to 20 percent by weight of UV-A absorbing agents which including an acid group, from ~~about~~ 0.25 to 20 percent by weight of UV-B absorbing agents which include an acid group, from 0.5 to 20 percent by weight of a fatty amine, from 50 to 90 percent by weight of water, from 1 to 20 percent by weight of an emollient, from 1 to 10 percent by weight of an emulsifier, from 0.05 to 2 percent by weight of preservatives and antioxidants, and less than 1 percent by weight of fragrances, coloring agents and chelating agents.

12. The formulation of claim 8, 9 or 10 comprising from 1 to 8 percent by weight of UV-A absorbing agents which include an acid group, from 1 to 8 percent by weight of UV-B absorbing agents which include an acid group, from 1

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to 8 percent by weight of a fatty amine, from 60 to 80 percent by weight of water, from 1 to 5 percent by weight of an emollient, from 1 to 5 percent by weight of an emulsifier, from 0.1 to 1 percent by weight of preservatives and antioxidants, and less than 1 percent by weight of fragrances, coloring agents and chelating agents.

13. The formulation of claim 8 in the form of a maximum protection sunblock comprising ~~about~~ 70 percent by weight of water ~~about~~ 4 percent by weight of 2-phenylbenzimidazole-5-sulfonic acid, ~~about~~ 4.5 percent by weight of triethanolamine, ~~about~~ 5 percent by weight of methoxycinnamic acid, ~~about~~ 2.5 percent by weight of salicylic acid, ~~about~~ 1 percent by weight of sulisobenzene, ~~about~~ 4 percent by weight of cocoyl imidazoline, ~~about~~ 3 percent by weight of ^{emollients,} ~~emollients,~~ ~~about~~ 5 percent by weight of emulsifiers, ~~about~~ 0.35 percent by weight of thickener, and up to 1 percent by weight of combined preservatives, antioxidants, fragrances and chelating agents.

14. The formulation of claim 8 in the form of a sunscreen which permits gradual tanning comprising ~~about~~ 80 percent by weight of water, ~~about~~ 4 percent by weight of 2-phenylbenzimidazole-5-sulfonic acid, ~~about~~ 1 percent by weight of sulisobenzene, ~~about~~ 6 percent by weight of cocoyl imidazoline, ~~about~~ 3 percent by weight of emollients, ~~about~~ 5 percent by weight of emulsifiers, ~~about~~ 0.35 percent by weight of thickener and up to ~~about~~ 1 percent by weight of combined preservatives, antioxidants, fragrances, and chelating agents.

15. The formulation of claim 7 comprising ~~about~~ 2 percent by weight of sulisobenzene, ~~about~~ 4 percent by weight of 2-phenylbenzimidazole-5-sulfonic acid, ~~about~~ 8 percent by weight of stearamidoethyl diethylamine, and ~~about~~ 86 percent by weight of ethanol.

16. A compound according to claim 1 substantially as



hereinbefore described with reference to any one of the examples.

17. A formulation according to claim 8 substantially as hereinbefore described with reference to any one of the examples.

DATED: 3 April, 1990

PHILLIPS ORMONDE & FITZPATRICK

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